

**Inferring the anthropogenic NO<sub>x</sub> emission trend over the United States during 2003 - 2017 from satellite observations: Was there a flattening of the emission trend after the Great Recession?**

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20 **Table S1. Summary of major satellite instruments for remote sensing of atmospheric NO<sub>2</sub> VCD in the past decade**

Instrument	Satellite	Launch date	End date	Operator	Equator crossing time (local time)	UV/Vis Spectral range (nm)	Spectral resolution (nm)	Swath length (km)	Nadir pixel resolution (km × km)	Global coverage (days)
SCIAMACHY	ENVISAT <sup>1</sup>	03/01/2002 <sup>2</sup>	04/08/2012 <sup>2</sup>	ESA <sup>3</sup>	10:00 <sup>1</sup>	240 – 805 <sup>4</sup>	0.24 – 0.48 <sup>4</sup>	960 <sup>5</sup>	60 × 30 <sup>5</sup>	6 <sup>5</sup>
GOME-2A	MetOp-A <sup>6</sup>	10/19/2006 <sup>6</sup>	in operation	EUMETSAT <sup>7</sup>	9:30 <sup>8</sup>	240 – 790 <sup>8</sup>	0.26 – 0.51 <sup>8</sup>	1920 before Jul. 15 <sup>th</sup> , 2013; 960 after Jul. 15 <sup>th</sup> , 2013 <sup>8</sup>	80 × 40 before Jul. 15 <sup>th</sup> , 2013; 40 × 40 after Jul. 15 <sup>th</sup> , 2013 <sup>8</sup>	1.5 <sup>9</sup>
GOME-2B	MetOp-B <sup>6</sup>	09/17/2012 <sup>6</sup>	In operation	EUMETSAT	9:30 <sup>8</sup>	240 – 790 <sup>8</sup>	0.26 – 0.51 <sup>8</sup>	1920 <sup>8</sup>	80 × 40 <sup>8</sup>	1.5 <sup>9</sup>
OMI	EOS-Aura <sup>10</sup>	07/152004 <sup>10</sup>	In operation	NASA	13:45 <sup>10</sup>	270 – 500 <sup>11</sup>	0.45 – 1.0 <sup>11</sup>	2600 <sup>11</sup>	24 × 13 <sup>11</sup>	1 <sup>11</sup>

21 <sup>1</sup> Refer to <https://earth.esa.int/web/guest/missions/esa-operational-eo-missions/envisat>  
22 <sup>2</sup> Refer to <https://en.wikipedia.org/wiki/Envisat>  
23 <sup>3</sup> The European Space Agency  
24 <sup>4</sup> Refer to <http://www.iup.uni-bremen.de/sciamachy/instrument/performance/index.html>  
25 <sup>5</sup> Refer to Boersma et al. (2008), Boersma et al. (2009), and Lee et al. (2009)  
26 <sup>6</sup> Refer to <https://www.eumetsat.int/website/home/Satellites/CurrentSatellites/Metop/index.html>  
27 <sup>7</sup> The European Organization for the Exploitation of Meteorological Satellites  
28 <sup>8</sup> Refer to EUMETSAT (2015)  
29 <sup>9</sup> Refer to Lee et al. (2009) and Wang et al. (2017)  
30 <sup>10</sup> Refer to <https://aura.gsfc.nasa.gov/>  
31 <sup>11</sup> Refer to <https://aura.gsfc.nasa.gov/omi.html>

32 **Table S2. Summary of satellite NO<sub>2</sub> TVCD products and their retrieval information**

NO <sub>2</sub> TVCD products	Version	Available period	DOAS fitting method	Stratosphere–troposphere separation	Fitting window (nm)	Albedo / reflectance	A priori profiles	Radiative transfer model	Cloud	Uncertainty
GOME-2B	TM4NO2A (2.3)	12/20/2012 – current	Intensity fit <sup>1</sup>	Assimilation of satellite total slant columns in the TM4 model <sup>2, 3</sup>	405 – 465 <sup>1</sup>	Climatology albedo from 3 years of OMI data <sup>4</sup>	TM4 (2° × 3°) <sup>2</sup>	DAK <sup>2</sup>	FRESCO+ (Oxygen A-band around 760 nm) <sup>5</sup>	1.0 × 10 <sup>15</sup> molecules/cm <sup>2</sup> + 25% <sup>2</sup>
SCIAMACHY	QA4ECV (v1.1)	08/02/2002 – 04/08/2012	Optical Density <sup>1, 6</sup>	Assimilation of OMI total slant columns in the TM5 - MP model <sup>6, 7</sup>	425 – 465 <sup>6</sup>	Climatology albedo based on SCIAMACHY <sup>8</sup>	TM5-MP (1° × 1°) <sup>6</sup>	DAK	FRESCO+	35% - 45% over polluted scenes; > 100% over background regions (Pacific Ocean) <sup>6</sup>
GOME-2A	QA4ECV (v1.1)	02/01/2007 – 12/31/2016			405 – 465 <sup>1, 6</sup>	Climatology albedo based on GOME-2A <sup>8</sup>			FRESCO+	
OMI-QA4ECV	QA4ECV (v1.1)	10/012004 – Current			405 – 465 <sup>1, 6</sup>	Climatology albedo from 5 years of OMI data <sup>6</sup>			Improved O <sub>2</sub> -O <sub>2</sub> (477 nm) <sup>9</sup>	
OMI-NASA	SPv3	01/01/2005 – 07/31/2017			402 – 465 <sup>1, 10</sup>	OMI climatology albedo <sup>10</sup>	GMI (1° × 1.25°) <sup>10</sup>	TMORAD <sup>10</sup>	O <sub>2</sub> -O <sub>2</sub> (477 nm) <sup>10, 11</sup>	SPv2.1 TVCD has uncertainties of about 30% under clear-sky conditions to about 60% under cloudy conditions <sup>12</sup> , and the relative difference between SPv3 and SPv2.1 is less than ~20% <sup>10</sup> .
OMI-BEHR <sup>13</sup>	v3.0B	01/01/2005 – 07/31/2017	Stepwise intensity fit with monthly averaged solar irradiance spectrum <sup>1, 10</sup>	Based on OMI total slant columns over regions with low estimated TVCD contributions (TVCD contributions less than 0.3 × 10 <sup>15</sup> molecules/cm <sup>2</sup> ) <sup>10</sup>		Based on MCD43D BRDF product (for land) and model parameterization (for ocean)	WRF-Chem (12 km)			

33 <sup>1</sup> Refer to Zara et al. (2018)

34 <sup>2</sup> Refer to Boersma et al. (2011). “TM4” is the Tracer Model, version 4. “DAK” is the Doubling-Adding KNMI (DAK) radiative transfer model.

35 <sup>3</sup> Refer to Williams et al. (2009)

36 <sup>4</sup> Refer to Kleipool et al. (2008)

37 <sup>5</sup> Refer to Wang et al. (2017) and Wang et al. (2008)

38 <sup>6</sup> Refer to Boersma et al. (2018)

39 <sup>7</sup> Refer to Williams et al. (2017)

40 <sup>8</sup> Refer to Tilstra et al. (2017)

41 <sup>9</sup> Refer to Veeffkind et al. (2016)

42 <sup>10</sup> Refer to Bucsela et al. (2013), Bucsela et al. (2016), Krotkov et al. (2017), and Marchenko et al. (2015). “TMORAD” is the TMOS radiative transfer model.

43 <sup>11</sup> Refer to Acarreta et al. (2004)

44 <sup>12</sup> Refer to Lamsal et al. (2014), Oetjen et al. (2013), and Tong et al. (2015)

45 <sup>13</sup> Refer to Laughner et al. (2018). OMI-BEHR uses the SCD from OMI-NASA SPv3 but updates inputs for the AMF calculation, such as a prior NO<sub>2</sub> vertical profiles and surface reflectance. Besides, OMI-BEHR only provides NO<sub>2</sub> TVCD over the contiguous

46 United States (CONUS). As in this study, we used the OMI-NASA datasets archived in the OMI-BEHR product, so we only obtained OMI-NASA datasets extended to July 31, 2017.

47 <sup>14</sup> Average uncertainty over the CONUS is calculated based on the file from <http://behr.cchem.berkeley.edu/behr/BEHR-us-uncertainty.hdf>

48 **Table S3. Selection criteria for satellite NO<sub>2</sub> TVCD pixel data**

NO <sub>2</sub> TVCD products	Period	Solar zenith angle	albedo	Cloud radiance fraction	Snow or ice covered	AMFtrop/AMFgeo	Flag for retrieval success	Retrieval quality flag	Rows in swath
GOME-2B	01/01/2013 – 12/31/2017	< 80°	<= 0.3	<= 50%	No	> 0.2	Yes		All
SCIAMACHY	01/01/2003 – 12/31/2011	< 80°	<= 0.3	<= 50%	No	> 0.2	Yes		All
GOME-2A	01/01/2008 – 12/31/2016	< 80°	<= 0.3	<= 50%	No	> 0.2	Yes		All
OMI-QA4ECV <sup>1</sup>	01/01/2005 – 12/31/2017	< 80°	<= 0.3	<= 50%	No	> 0.2	Yes		6 - 21
OMI-NASA <sup>1</sup>	01/01/2005 – 12/31/2016	< 80°	<= 0.3	<= 50%			Yes	Yes	6 – 21
OMI-BEHR <sup>1</sup>	01/01/2005 – 12/31/2016	< 80°	<= 0.3	<= 50%			Yes	Yes	6 - 21

49 <sup>1</sup> Rows 6-21 are selected to remove the anomalies developed in the OMI sensor (Boersma et al., 2018; Zhang et al., 2018).

50 **Table S4. Summary of annual trends of AQS NO<sub>2</sub> surface concentrations and satellite NO<sub>2</sub> TVCD products in each region during different periods<sup>1</sup>**

		Northeast		Midwest		South		West	
		AQS site	CONUS	AQS site	CONUS	AQS site	CONUS	AQS site	CONUS
AQS NO <sub>2</sub> VMR at 13:00 -14:00	2003 – 2011	-6.8 ± 0.7%		-6.1 ± 1.2%		-6.6 ± 0.7%		-7.6 ± 1.2%	
	2011 – 2017	-8.0 ± 1.2%		-6.4 ± 0.8%		-5.8 ± 0.6%		-7.2 ± 1.6%	
AQS NO <sub>2</sub> VMR at 10:00 – 11:00	2003 – 2011	-6.6 ± 0.5%		-5.8 ± 1.5%		-6.5 ± 1.3%		-7.1 ± 1.6%	
	2011 – 2017	-7.6 ± 1.0%		-6.8 ± 0.5%		-5.7 ± 0.1%		-6.1 ± 1.1%	
SCIAMACHY	2003 – 2011	-17.1 ± 2.7%	-11.0 ± 3.3%	-12.9 ± 6.8%	-6.5 ± 0.8%	-9.1 ± 1.0%	-6.2 ± 1.5%	-9.1 ± 1.8%	-7.0 ± 1.4%
	2011 – 2017								
GOME2B	2003 – 2011								
	2013 – 2017	-11.4 ± 3.7%	-10.8 ± 3.9%	-9.9 ± 13.1%	-4.4 ± 27.2%	-8.9 ± 3.0%	-7.5 ± 3.6%	-11.8 ± 3.0%	-10.6 ± 2.3%
OMI-QA4ECV	2005 – 2011	-14.2 ± 6.3%	-10.6 ± 3.8%	-9.2 ± 4.2%	-8.4 ± 2.8%	-9.2 ± 2.7%	-8.2 ± 1.5%	-10.5 ± 1.6%	-8.7 ± 0.9%
	2011 – 2017	-18.0 ± 16.2%	-7.6 ± 4.2%	-7.6 ± 3.3%	-7.0 ± 1.7%	-4.8 ± 1.4%	-4.6 ± 1.0%	-6.4 ± 1.4%	-4.8 ± 1.2%
OMI-NASA	2005 – 2011	-11.8 ± 1.3%	-11.0 ± 1.8%	-10.9 ± 4.8%	-10.0 ± 4.1%	-10.0 ± 3.5%	-9.5 ± 1.9%	-10.2 ± 1.8%	-8.5 ± 0.9%
	2011 – 2016	-10.0 ± 4.9%	-8.5 ± 3.8%	-13.2 ± 3.2%	-9.2 ± 2.7%	0.3 ± 19.2%	-8.0 ± 5.5%	-9.0 ± 5.7%	-6.6 ± 3.9%
OMI-BEHR	2005 – 2011	-11.8 ± 1.8%	-10.9 ± 1.9%	-12.2 ± 7.3%	-9.8 ± 4.4%	-9.5 ± 3.1%	-8.8 ± 2.0%	-9.9 ± 1.1%	-8.2 ± 0.4%
	2011 – 2016	-8.2 ± 3.4%	-6.6 ± 1.7%	-27.4 ± 24.3%	-8.1 ± 3.0%	-7.2 ± 2.3%	-5.0 ± 1.3%	-13.2 ± 14.5%	-7.0 ± 4.8%

51 <sup>1</sup> Annual trends are the averages of regional seasonal trends (e.g, Figure 7).

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